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TOPICAL REVIEW

Why are atomic and molecular dimers so exciting?

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Abstract

The large number of experimental and theoretical studies devoted to atomic and molecular van der Waals dimers is here to attest for the strong interest and excitement raised in the scientific community over the past decade. One of their intriguing feature is the ability of the two constituting monomers to interact at incredibly large distances through newly evidenced processes, such as Interatomic Coulombic Decay, due to the transition from closed to open shells upon ionization. Beyond the interest for their intrinsic properties, dimers are also valuable for the large amount of information obtained on the monomers themselves, in particular in the case of collisions with ions. Indeed, one of the first striking findings was that dimers behave as two quasi-independent monomers regarding collision processes. This result triggered a series of experiments highlighting specific properties of atomic and molecular dimers, viewed alternatively as new species or as monomers in a minimal environment. They concern several fields, from electron emission to molecular fragmentation. This review presents the diverse and surprising facets of atomic and molecular dimers uncovered through ion collision experiments over the past fifteen years.

1. Introduction

Since the discovery in 1997 of an unexpected electronic decay mechanism occurring in weakly bound systems by *et al* [1], van der Waals dimers have focused much attention through experimental as well as theoretical studies. To date, this process, known as Interatomic Coulombic Decay (ICD), has been the subject of more than 300 articles and more than 10 review papers [2 and references therein]. The dimer excited states leading to ICD, usually described as a non-local Auger–Meitner decay, are efficiently produced through resonant photoionization, which explains why most of the experiments have been performed at synchrotron radiation facilities.

Ion beams were introduced about 10 years later using van der Waals dimers as targets. The initial motivation was their distinct properties compared to molecules, with a reduced number of potential energy curves (and curve crossings), and the resulting impact on charge rearrangement prior to fragmentation [3]. But the harvest of knowledge was much larger than expected. The potential of van der Waals dimers when used as targets revealed several aspects of the collisions unreachable with monomer targets. These unique advantages of dimers in ion–atom and ion–molecule collision physics (and beyond) will be presented and explored in the following.

After the first experimental measurement of the ICD process, several other non-local processes have been either evidenced such as Electron Transfer Mediated Decay or predicted such as Interatomic Coulombic Electron Capture. This general non-local character of electronic decay processes has attracted a significant interest from the scientific community on these weakly bound systems.

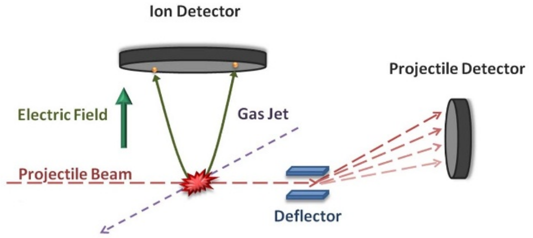

Figure 1. COLTRIMS spectrometer configuration showing an electrostatic analyzer to determine the projectile charge state. Reprinted from [8], Copyright (2003), with permission from Elsevier.

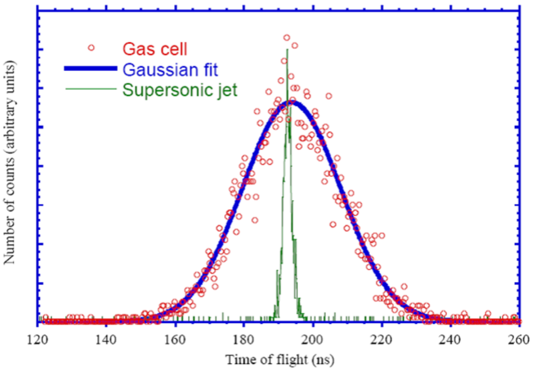

Figure 2. Ar^+ time-of-flight peaks measured in a collision with a 6.7 MeV/u Ar16+ projectile when the target is a gas cell (red circle) or a supersonic gas jet (green line). Reproduced with permission from [9].

2. Experimental technique

Measuring the momentum of the particles resulting from a collision is a long standing technique in nuclear physics to access to the collision impact parameter or, since in most of the cases the collision is inelastic, to measure the internal energy change during the collision, which is known as the reaction Q-value. This is the driving idea for the development of the COLd Target Recoil Ion Momentum Spectroscopy (COLTRIMS) technique having in mind its application to atomic and molecular collisions.

The COLTRIMS has been invented for ion–atom collision studies at the end of the 80's. Three pioneering groups (Kansas State University, Frankfurt University and CIMAP laboratory) have introduced coincidence and imaging techniques in their experimental setup. The dynamics of the collision as well as all correlations became directly accessible experimentally. This was the birth of a kinematically complete technique for atomic and molecular collision physics giving access to the full dynamics of such quantum systems [4–7].

The momentum measurement relies on the extraction of the ionized target recoil, being either atomic ions or molecular ion fragments, by an electric field surrounding the collision region and detected by a position sensitive detector (figure 1). Position and time of flight measurements in coincidence lead to the determination of the full momentum vector of the recoil ion. The resolution is fixed by both the initial velocity distribution in the target gas jet and the target gas jet diameter. Both quantities are suitably provided by a supersonic expansion gas jet. Furthermore, COLTRIMS provides a 4π solid angle collection efficiency.

The device providing target atoms is the key point of the COLTRIMS technique which needs a narrow enough momentum distribution to address the very weak momentum transfers occurring during an ion–atom collision. A standard gas cell at room temperature has a Boltzmann momentum spread more than ten times larger than the recoil momentum to be measured (figure 2). The solution came from the use of a supersonic expansion which provides atoms with a narrow velocity distribution. Their mean kinetic energy is nearly twice the thermal Boltzmann mean value but the width of the momentum distribution is of the order of ± 0.05 a.u. along the beam direction and ± 0.035 a.u. in the transverse direction [9] (figure 2). This low temperature of the jet leads also to the production of dimers when properly tuned. The typical diameter at the collision point is of the order of 1 mm providing the necessary position measurement resolution. The jet density can be as high as 10^{12} atoms/cm^3 in the collision volume.

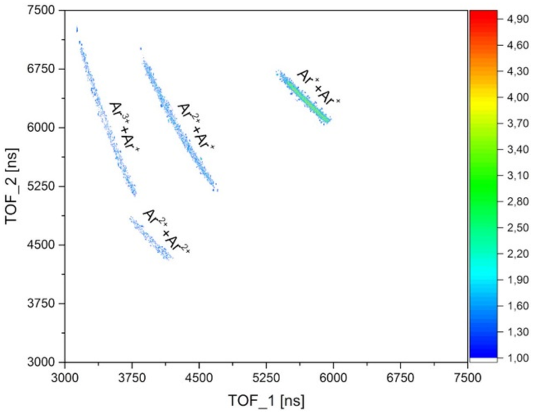

Figure 3. Coincidence map for Ar_2 dimer fragmentation after background filtering using momentum conservation.

In the case of molecule or dimer targets, momentum conservation may be applied to the complete fragmentation channels, corresponding to the detection of all fragments. By definition, the sum of the fragments momentum is equal to zero in the center-of-mass frame. In the case of two-body dissociation, this implies that the fragments are emitted back-to-back. The following experiments exploit this principle of momentum conservation among the dimer fragments. The results are an improvement of the momentum resolution, a better separation between all the dissociation channels and a high cancellation of the background counts due to random coincidences rejection [10] (figure 3). This powerful technique has been the key factor in revealing the results presented in this paper.

3. Dimers as new target species

Collisions between multiply charged ions and atoms or molecules have been extensively studied for many years, and the induced electronic processes such as excitation, ionization [11] and capture [12] are now well understood. In the case of molecules, and when several electrons are removed from the target, the collision is essentially followed by its fragmentation [13]. For diatomic molecules, it has been shown (i.e. [14, 15]) that a preferential charge symmetric fragmentation occurs. This preference is strongly linked to the fact that in molecules, valence electrons are delocalized over the whole molecule, which could be pictured as a high electron mobility across the molecule. This covalent character is the reason for the large binding energy and the short internuclear distance, 9.87 eV and 1.1 Å, respectively, for N_2. The situation is completely different for rare-gas dimers. In these systems, the ground state valence orbitals are localized on each atomic center, resulting in larger interatomic separation and lower binding energy, 3.8 Å and 12 meV, respectively, for Ar_2 dimers. These specific characteristics make rare-gas dimers a unique class of weakly bound species which may behave differently than molecules, notably regarding the electronic arrangement around the fragments.

In the case of double electron capture from Ar_2 by a multiply charged ion such as Ar^{9+} at 152 keV, the electrons may be captured either from the two atomic sites (two-site capture) or from one single Ar atom (one-site capture) [3]. For the one-site capture, measurements of the kinetic energy released (KER) by the dimer dication fragmentation revealed the occurrence of the radiative charge transfer (RCT) process. This slow relaxation process requiring photon emission is the only way for the $[Ar^{2+}–Ar^{0}]$ electronic state to de-excite to the dimer dication ground state. It is the signature of the low electron mobility across the dimer. RCT can be seen as a memory effect which allows to discriminate *a posteriori* between one-site and two-site electron removal.

The same conclusion on electron mobility can be derived from the four-fold electron removal. In that case, two channels involving charged fragments are available. The symmetric channel $[Ar^{2+}–Ar^{2+}]$ and the asymmetric one $[Ar^{3+}–Ar^{+}]$. For molecules such as N_2 or CO, the asymmetric channel represents around 20% of the symmetric channel. For Ar_2 dimers (figure 4), the asymmetric channel strongly increases to reach 2.45 times the symmetric channel, showing a strong preference for the asymmetric fragmentation channel [16].

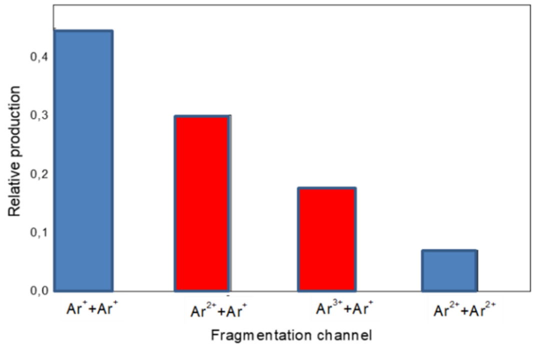

Figure 4. Relative yield of the different fragmentation channels obtained in the collision between Ar^{9+} projectile ions at 152 keV and Ar_2 dimers. Reprinted figure with permission from [16], Copyright (2018) by the American Physical Society.

This first example shows how the distinct characteristics between atomic dimers and covalent molecules lead to a completely different behaviors regarding relaxation and fragmentation processes following ion impact. The low electron mobility observed for dimers allows to keep memory of the electron removal configuration (one-site vs two-sites). The very weak interaction between the two monomers and the resulting low electron mobility are two characteristics which suggest that the dimer may be seen as two quasi-independent atoms fixed in space. It is equivalent to say that the dimer electron density is treated as the simple superposition of the two monomer electron densities. Thus, this is the simplest representation of an atom surrounded by an environment made of another single atom. The following chapters will show what can be learned from this picture.

4. Role of the environment

Atom and molecule interactions with radiation are studied experimentally since many years [13, 17]. The target is generally in the gas-phase in order to ensure that the projectile interacts with a single and isolated monomer. If an atom or a molecule is sitting near i.e. (at a distance comparable to relevant impact parameters) the target atom or target molecule, then the target is in the presence of what is usually called an environment, weakly interacting as in liquids. A dimer provides the simplest form of environment with one atom (or molecule) being considered as the target and the second atom (or molecule) as the environment. Then, in the presence of such an environment, many questions arise: are the collision processes modified compared to the case of an isolated monomer (atom or molecule)? Do new processes appear due to the surrounding species? In the last years, the tendency was to study more and more complex molecules which are commonly found in biology. An important question is how relevant to biology are the results obtained from the gas-phase? In more realistic conditions, such as those found in living organisms, what is the influence of the surrounding environment on the collision? The dimers offer a unique opportunity to investigate this influence, while keeping access to fragmentation dynamics measurements.

4.1. Is a dimer equivalent to two independent monomers?

4.1.1. Atomic dimers

One approach to probe the environment effects on collision processes is to compare experimental results to model calculations assuming no direct interaction between the target and its environment. In the case of atomic dimers, experimental results obtained in collisions between highly charged ion projectiles and rare gas dimer were compared to a model assuming two independent atoms sitting next to each other at the equilibrium interatomic distance of the dimer. The observables of interest are the relative yields of the different fragmentation channels produced during the collision (figure 5), as well as the associated distributions in scattering angle. The calculation is based on the extended over-the-barrier model [18] combined with a Monte-Carlo code in order to simulate the different dimer orientations and impact parameters needed to generate the different cross sections [16].

This comparison has been analyzed for two different projectiles (Ar^{9+}, Xe^{20+}) and two different dimers (Ar_2, Ne_2). These two projectiles have been chosen to compare medium charged projectiles to highly charged projectiles.

The relative yields obtained for Ar^{9+} projectiles impinging on Ar_2 dimers are presented in figure 5. For all four collision systems, a fairly good agreement of the calculation with experimental data was

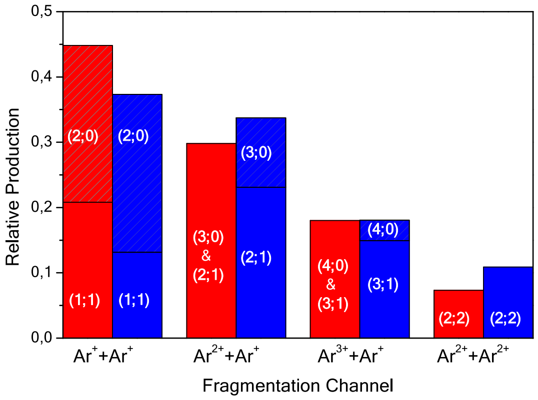

Figure 5. Experimental (red) and calculated (blue) relative yields of the different fragmentation channels obtained in the collision between Ar^{9+} projectile ions at 152 keV and Ar_2 dimers. Reprinted figure with permission from [16], Copyright (2018) by the American Physical Society. The initial charge states of the fragments contributing to the final fragmentation channel are noted in brackets (i.e. (3;1)).

observed both for relative yields and scattering angle distributions [16]. The clear success of this atomic model shows that regarding the energetic collision process, rare gas van der Waals dimers can be well represented as two independent atoms.

The first important conclusion which can be derived from these observations is that the energetic collision atomic processes, here the electron capture (single and multiple), are weakly affected by the presence of the environment.

4.1.2. Molecular dimers

Ion induced molecular fragmentation is usually assumed to be a two-step process. The first step corresponds to the removal of electrons from the molecule by the interaction process with the projectile ion, which can be either ionization or electron capture. It corresponds to a transition from the neutral molecule ground state to an excited state of the resulting molecular ion. This transition is assumed to happen without any nuclei motion (vertical transition), as the interaction time, around 200 attoseconds or less, is much shorter than the typical vibration period of the molecule. The molecular ion excited state is generally dissociative and de-excites through fragmentation. The resulting KER spectrum provides an original spectroscopic tool for the study of dissociative molecular ions, as it corresponds to the potential energy difference between the excited state and the final state of the separated fragments. This spectrum is the signature of the interaction between the projectile and the monomer (figure 6(a)). In order to reveal the role of the environment on the molecular fragmentation process, collisions between $(N_2)_2$ dimers and 90 keV Ar^{9+}[19] were investigated, focusing on the KER obtained for the fragmentation channel $(N_2)_2{}^{3+} \rightarrow N_2{}^{+} + N^{+} + N^{+}$. The idea was to measure the fragmentation KER of one of the N_2 molecule when the other one does not dissociate. Figure 6(b)) shows the superposition of the dimer KER spectrum with the one of the isolated molecule, shifted by 6.7 eV. This energy shift corresponds to the additional potential energy resulting from the repulsive interaction between the N_2^+ fragment and the two N^+ ions. Quite surprisingly, these two curves were found to be very similar, evidencing that the same excited states of the molecular ion are populated during the collision with comparable relative cross sections.

The second important conclusion which can be derived here for molecules is the weak influence of the environment on the primary processes, in this case the electron capture (single and multiple), and on the subsequent molecular fragmentation.

4.2. Does the environment play a role?

The previously described experiments show that collision processes are not significantly affected by the presence of an environment. This is true for the single and multiple electron capture cross sections, as observed on atomic dimers. This is also true for the cross sections associated to the molecular ion excited states, as shown by the KER spectra of molecular dimers. The remaining question is then, does the environment have no influence at all or can it give rise to new processes, which are not present for an isolated molecule?

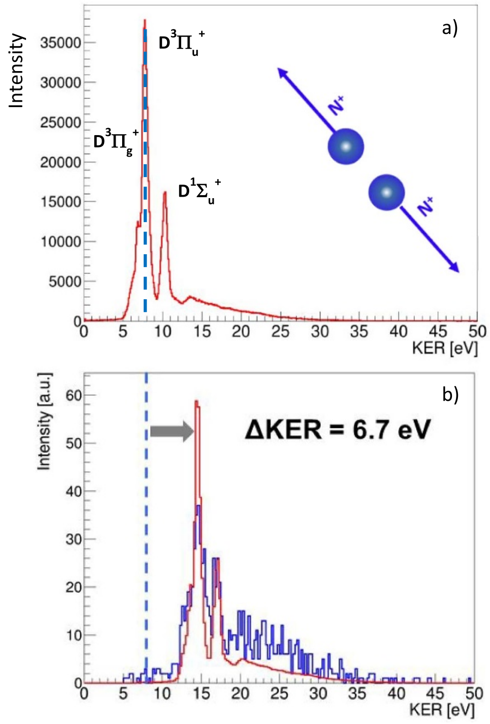

Figure 6. (a) Experimental kinetic energy release spectrum for the dication fragmentation channel $N_2^{2+} \rightarrow N^+ + N^+$ resulting from the collision of a 90 keV Ar^{9+} projectile ion and an N_2 molecule. Reprinted figure with permission from [19], Copyright (2017) by the American Physical Society. The dotted blue line corresponds to the position of the main peak of the dication KER distribution. (b) Experimental KER spectrum for the dication fragmentation channel $N_2^{2+} \rightarrow N^+ + N^+$ (red) and the trication fragmentation channel $N_2^{3+} \rightarrow N_2{}^+ + N^+ + N^+$ (blue) resulting from the collision of a 90 keV Ar^{9+} projectile ion and an N_2 molecule and N_2 dimer respectively. The dication spectrum is shifted by 6.7 eV. The dotted blue line corresponds to the position of the main peak of the dication KER distribution prior to the shift.

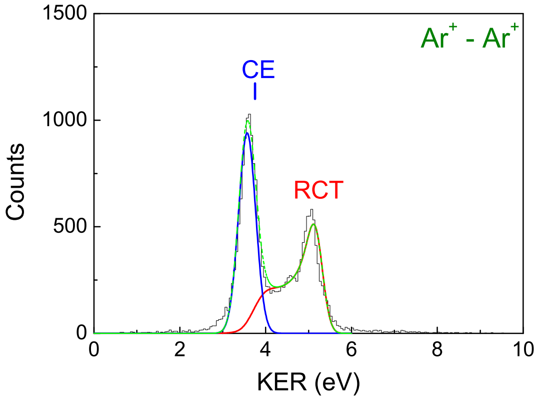

Figure 7. Experimental KER spectrum for the Ar_2^{2+} fragmentation channel $Ar_2^{2+} \rightarrow Ar^+ + Ar^+$. Two processes, Coulomb explosion and Radiative Charge Transfer are identified. Reprinted figure with permission from [3], Copyright (2010) by the American Physical Society.

4.2.1. RCT

As mentioned earlier in section 3 for the case of Ar_2 dimers, RCT is the process allowing [Ar^{2+} $2p^{-2}$–Ar^0] electronic states to de-excite to the dimer dication ground state. Due to the polarization attraction, these states have a potential energy surface with a minimum at a shorter internuclear distance (3 Å) than the 3.8 Å equilibrium internuclear distance of the neutral dimer. Their radiative lifetime (∼ns) are much longer than the vibrational period (∼hundreds of fs). The dimer has then sufficient time to undergo vibrational motion and de-excite through a radiative transition to the [Ar^+–Ar^+] state close to the inner turning point, thus leading to a higher KER (figure 7) [3].

The RCT process can occur only when a second atom is in the vicinity of the dication produced during the collision. This is a clear consequence of the presence of an environment around the target atom.

4.2.2. ICD

This non-local ICD process, originally predicted by theory [1] and observed experimentally for the first time in the 00's [20, 21], has renewed the interest in van der Waals dimers, triggering a large number of studies on dimer interaction with radiation (photons, electrons, ions) [2]. After production of an inner-shell vacancy in one of the dimer's monomer (atom or molecule), the energy resulting from a valence electron filling this vacancy is transferred to the neighbor monomer, where a low energy secondary electron is ejected. Whereas ICD was evidenced in high-energy ion collisions [22], this process was unexpected in low-energy multiply charged ion collisions. It was nonetheless recently evidenced with Ne_2 dimers [23].

The ICD process can occur only when a second monomer is next to the monomer holding an inner-shell vacancy. ICD is thus a specific signature of the presence of an environment around the excited target.

4.2.3. Conclusion

The appearance of these two processes, RCT and ICD, clearly shows that the environment has an effect on the relaxation of the excited monomer.

RCT causes the dimer implosion by shrinking the inter-nuclear distance before the Coulomb explosion. The resulting larger KER value is the key to recover the memory of the electron removal configuration (one-site vs two-sites).

Apart from direct ionization or Auger–Meitner cascades, ICD appears as a new source of electrons in ion-matter interaction [23]. This new finding is particularly important, as it occurs near the stopping point of the ion in matter, increasing the amount of energy deposited at the location of the Bragg peak.

5. Input from a 'spectator'

Certain physical quantities are difficult to access experimentally when dealing with isolated monomer targets. The two following examples, one concerning atoms and the other molecules, demonstrate how helpful and powerful is the presence of a 'spectator' monomer, as found in a dimer, standing next to the target monomer.

5.1. Atomic dimers

In ion–atom collisions, the ionization probabilities are usually calculated as a function of the impact parameter b. The COLTRIMS technique was originally developed to access this parameter b in high-energy collisions, where its determination through the measurement of the projectile scattering angle is not possible anymore, due to its very small value. It became very soon evident that in such ionizing collisions, the recoil ion momentum does not solely reflect the impact parameter because it is also balancing the ejected electron momentum, as in photoionization processes [24, 25]. The momentum transfer from the projectile to the target is then hidden by the momentum carried out by the electrons. However, it has been shown for diatomic molecules that in some cases, the ionization cross-sections depend on the angle between the molecular axis and the ion beam axis. These orientation dependent cross sections are influenced by the impact parameter-dependent ionization probabilities P(b) when the molecule interatomic distance is in the range of the active impact parameters [26, 27]. This interatomic distance effectively acts as a ruler for impact parameter dependent measurements.

In a dimer, the presence of a 'spectator' monomer adds another dimension to the spherical symmetry of the atom, allowing dimer orientation dependent measurements, as discussed previously for covalent diatomic molecules. This strategy was applied successfully to He_2 and Ne_2 dimers [28, 29]. A good agreement was obtained between the simulation of orientation effects, using theoretical impact parameter dependent probabilities, and the experimental observations (figure 8).

Finally, it is clear that benchmarking theoretical impact parameter dependent probabilities, unreachable experimentally in ion–atom collisions, becomes possible thanks to the presence of the 'spectator' atom available in a dimer, at a distance compatible with the dominant range of impact parameter. A similar strategy has been successfully applied in low energy collisions as well, to obtain the impact parameter dependent electron capture probabilities. A strong preference for electron capture by the projectile from the nearest site of the dimer was evidenced [30].

5.2. Molecular dimers

During a collision, the dication molecular excited states that are produced are generally dissociative. Some of these excited states may be metastable and the question of their identification arises. Diatomic

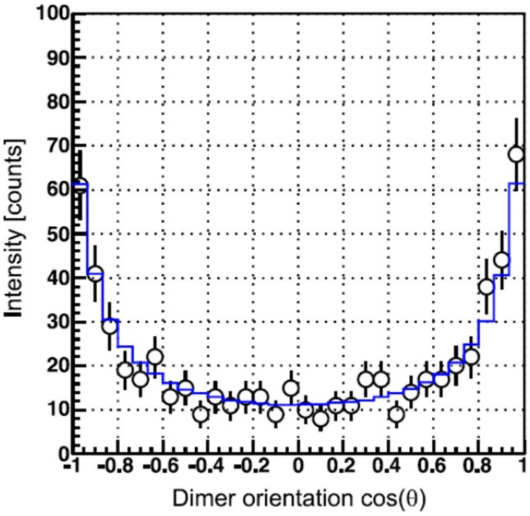

Figure 8. Relative cross sections as a function of the dimer orientation for the fragmentation channel $He^+ + He^+$ in the case of $S^{14+} + He_2$ collisions. Reprinted figure with permission from [29], Copyright (2014) by the American Physical Society.

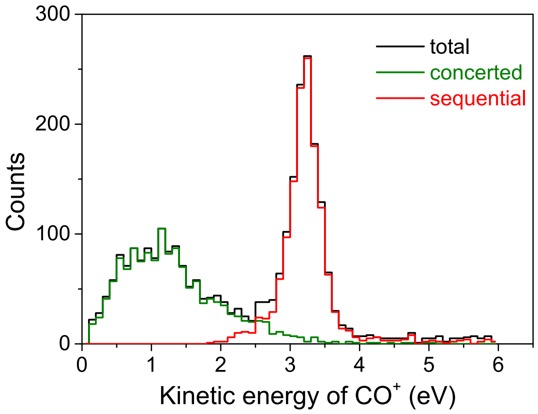

Figure 9. Kinetic energy of the CO^+ fragment for the fragmentation channel $[(CO)_2{}^{3+} \rightarrow CO^+ + C^+ + O^+]$. Reprinted figure with permission from [31], Copyright (2021) by the American Physical Society.

molecular ion fragmentation dynamics is a simple two-body dissociation which does not allow for discrimination between metastable or dissociative excited states. An elegant way to overcome this limitation is to produce these excited states within a molecular dimer and study their dissociation. In this configuration, the fragmentation dynamics will be very different depending on whether the molecular ion dissociates prior to, or during, dimer dissociation.

The study of $(CO)_2$ dimers is a very good example to illustrate the fragmentation dynamics of the CO^{2+} dication [31]. The dimer trication may dissociate either in a sequential way, through the channel $[(CO)_2{}^{3+} \rightarrow CO^+ + CO^{2+} \rightarrow CO^+ + C^+ + O^+]$, or in a concerted way, through the dissociation channel $[(CO)_2{}^{3+} \rightarrow CO^+ + C^+ + O^+]$. The sequential fragmentation channel is associated to a metastable state of the CO^{2+} dication, whereas the concerted fragmentation one, to a dissociative state of CO^{2+}. The kinetic energy of the 'spectator' molecular ion, CO+, is the key to unveil the metastability of the dication excited state. For a metastable state, the 'spectator' CO^+ is emitted within a two-body breakup, with a kinetic energy resulting simply from the Coulomb repulsion with the metastable CO^{2+} molecular ion. For a dissociative state of CO^{2+}, the kinetic energy of the 'spectator' CO^+ results from the Coulomb repulsion within a three-body breakup, where the two other singly charged fragments, C^+ and O^+, are rapidly moving away. The kinetic energy of the CO^+ 'spectator' is then smaller than in the former case of a metastable CO^{2+} dication excited state. This is clearly shown in figure 9 where the two fragmentation dynamics appear as two well separated peaks in CO^+ kinetic energy.

The fragmentation dynamics associated to these peaks was also confirmed by plotting the corresponding Newton diagram and Dalitz plot [31]. Even more interestingly, the KER spectrum corresponding to each peak indicates directly the states which are metastable and those which are not (figure 10). It

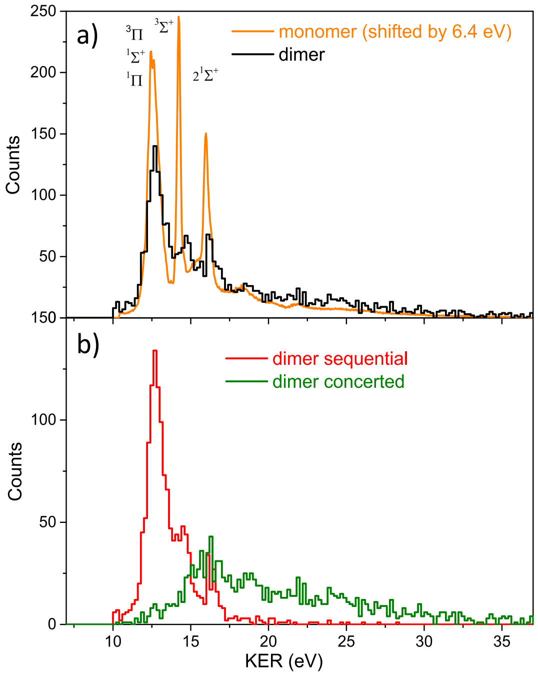

Figure 10. (a) Total KER spectrum for the $(CO)_2^{3+}$ trication dissociation in $[CO^+ + C^+ + O^+]$ (black) and for the CO^{2+} dication dissociation in $[C^+ + O^+]$ (orange) normalized to dimers and shifted by 6.4 eV (see § 4.1.2). (b) KER spectrum for sequential fragmentation (red) and concerted fragmentation (green). Reprinted figure with permission from [31], Copyright (2021) by the American Physical Society.

appears that metastable states correspond to low KER dissociation peaks, associated to the lowest excited states of CO^{2+} dication. The characteristic time of the fragmentation makes this technique well suited to identify metastable states with a lifetime longer than about 100 fs.

These results show how helpful can be the 'spectator' in deriving important information on the monomer itself by carrying the finger prints of the monomer fragmentation process.

6. Conclusion

The goal of this paper is to highlight the huge amount of new knowledge gained from studying these surprisingly rich and seemingly simple dimer targets. These systems made of two weakly interacting monomers, either atoms or molecules bound together but without any share of electrons, provide an intermediate situation between isolated atoms and covalent molecules. Their interaction with ions can be treated independently, as far as the initial collision processes are concerned, such as excitation, ionization or electron capture.

At the same time, they can also be viewed as representing a monomer surrounded by the simplest possible weakly interacting environment. This situation corresponds to an 'elemental' condensed-phase analog, allowing for the application of high-resolution and coincident techniques such as COLTRIMS, giving access to detailed analysis of environment effects. We have shown that whereas the environment has sometimes an influence and sometimes not, two main conclusions can be drawn. The first one is that the environment does not play an important role on the collision processes during the projectile-monomer interaction, for which each monomer behaves as if it was isolated. However, and this is the second conclusion, the relaxation of the excited monomer does depend strongly on the presence of the neighbor monomer. In particular, new processes such as RCT and ICD, may occur, due to this surrounding. This picture agrees with the separation of the physical collisional effects from the physico-chemical post-collisional effects made in solute/solvent interaction numerical simulations, as for example in the case of radiolysis [32, 33]. Indeed, the first step treats the ion interaction with isolated monomers and is called 'physical step'. Then, the relaxation of the excited monomer is treated taking into account the environment and corresponds to the 'chemical step'.

The attached monomer can also be viewed as a spectator providing information not accessible from the monomer alone, such as the collision impact parameter or the lifetime of a molecular ion excited state.

The dimer story is far from complete as many exciting questions are ahead. For instance, the precise geometry of certain molecular dimers is not fully determined and i.e. $(CO)_2$ dimers are still challenging on this point [34]. But the widely open field of research is the study of chemical reactions within molecular dimers, which is in its beginning [35]. On the theoretical side, awaited calculations are under development and will be of great help to fully understand the ion dimer interaction [36].

Data availability statement

The data cannot be made publicly available upon publication because no suitable repository exists for hosting data in this field of study. The data that support the findings of this study are available upon reasonable request from the authors.

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